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# **Influence of subsurface cracks on laser induced surface damage**

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## **Summary**

Cracks can affect laser damage susceptibility in three ways. These are field intensification due to interference, enhanced absorption due to trapped material in the cracks, and increased mechanical weakness. Enhanced absorption is the most important effect.

## **Introduction**

From the beginning of studies of laser-induced damage to optics over three decades ago, researchers have noted a correlation<sup>1</sup> between properties of the polishing caused sub-surface mechanically damaged layer and the susceptibility to laser induced surface damage. However, the precise nature of the connection is not always clear since cracks can influence each of the three necessary conditions for macroscopic damage. These conditions are 1) adequate energy density stored in the laser beam, 2) an absorption mechanism to couple laser energy into the optical material and 3) mechanical weakness leading to irreversible changes and fracture in the material. Further, cracks can potentially affect the thresholds of both intrinsic and extrinsic damage mechanisms. Early theoretical studies<sup>2</sup> were made of the intensification inside cracks due to electromagnetic boundary conditions. Recent studies<sup>3</sup> have attempted to relate the transition from plastic deformation to brittle fracture behavior of cracks with laser-induced damage. The use of advanced finishing techniques<sup>4</sup>, which reduce surface normal loads during polishing, reduce the amount of subsurface mechanical damage and increase the laser damage hardness.

The purpose of this report is to describe the ways in which near surface cracks can affect laser-induced damage susceptibility. It is organized according to the three necessary conditions for macroscopic damage listed above. Laser beam intensification due to either electromagnetic boundary conditions at crack walls or to total internal reflection at cracks and free surfaces along with interference increases the available light energy density. Particulate matter, especially that derived from polishing, can be trapped in cracks. Together with chemical bond changes at crack walls, such trapping adds absorbing centers near the surface. Finally, the presence of cracks mechanically weakens the material making it easier to cause macroscopic damage for given energy input. We consider both intrinsic (dielectric breakdown) and extrinsic (absorption by small particles) mechanisms, details of which are given in Appendices 1 and 2. We find that enhanced absorption due to either particulate matter trapped in cracks or clusters of oxygen deficient chips formed during crack formation are the most likely source of the crack related damage enhancement in fused silica surface damage.

## **Field intensification by cracks**

Field intensification in cracks was first discussed by Bloembergen<sup>2</sup> based on a very simple model. Consider the crack as a thin slit with width much smaller than the laser wavelength. In this situation, the field inside

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the crack can be treated as electrostatic. Consider the component of electric field normal to the crack. From the boundary conditions, the relationship between the field in the material  $E_0$  and the field inside the crack  $E_I$  is given by

$$E_I = \epsilon E_0 \quad (1)$$

where  $\epsilon = n^2$  is the dielectric constant of the material. For fused silica with refractive index  $n=1.5$ , the intensity inside the crack can be  $n^4 \sim 5$  times higher than in the bulk material. For typical laser parameters, this amount of intensification is not sufficient to produce intrinsic damage (see Appendix I). The principal problem with this argument, however, is that the intensification is only large for empty cracks. If the crack is filled by some material with refractive index  $n_f$ , the intensification drops  $n_f^4$  times.

A more interesting type of intensification was suggested<sup>5</sup> by Génin et al. The crack can reflect laser light that then interferes with the main beam causing intensity hot spots. Multiple reflections from cracks with the right orientation and the rear surface are particularly effective since total internal reflection can occur.

Consider intensification induced by a halfpenny crack of size  $a$  shown schematically in Fig. (1). The most dangerous situation takes place when the crack totally reflects incident

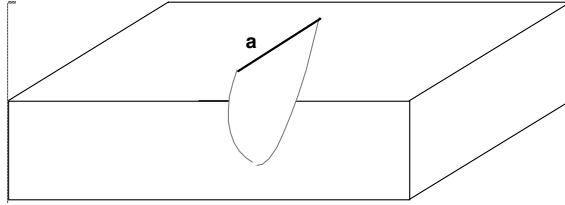


Fig. 1: Schematic halfpenny crack with diameter  $a$ . Crack opening assumed wide enough to permit total internal reflection.

light toward the rear surface. Then, this light totally reflects from the surface and interferes with the incident beam producing field intensification up to three times (intensity increases 9 times). The field near the surface for S polarization is, in this case,

$$E = E_0(1 + 2\cos(k_t x)) ; \quad \text{intensity} \propto E^2 \quad (2)$$

where  $k_t$  is the component of wavenumber parallel to the rear surface.

The regions of light intensification on the surface are parallel strips of length  $a$ . This intensification is still too low to reach intrinsic damage thresholds. Aside from the estimates of the intrinsic threshold given in Appendix 1, it should be noted that the intrinsic mechanism depends only on laser intensity so the intrinsic damage threshold fluence will scale as the pulselength. This contradicts observation and is further evidence that the actual damage mechanism is extrinsic. But field intensification can enhance the damage induced by microinclusions (see Appendix 2). To estimate the overall importance of this effect we must calculate the damage enhancement factor (DEF), which we now describe.

We introduced the concept of damage enhancement factor earlier to quantitatively assess the effect of beam modulation (e.g. caused by diffractive optics) on initiation of laser damage. When surface damage is due to an underlying distribution of extrinsic initiators, it can be characterized by the cumulative density  $c(F)$ , which is the number of initiators per unit area that initiate at fluence  $F$  or less. Typically,  $c(F)$  is a strong function of laser fluence, e.g. a power law or exponential function. Beam hot spots then cause disproportionately more damage than expected from the intensity distribution. However, if the area over which the hot spot occurs is small, it may still be unimportant.

Suppose  $N$  is the number of damage sites initiated over area  $S$  by the fluence distribution  $F(x,y)$ . Then we can write

$$N = \int_S c(F(x,y)) \, dxdy \equiv c(F_{eff})S \quad (3)$$

which defines  $F_{eff}$  as the effective uniform fluence that would cause the same amount of damage as the actual fluence distribution. The relative increase in number of damage sites over that which would be initiated at constant fluence  $F_0$  is given by the damage enhancement factor (DEF)

$$DEF = \frac{N}{N_0} - 1 = \frac{c(F_{eff})}{c(F_0)} - 1 \quad (4)$$

In the case of a crack network, if fluence  $F(x,y)$  is due to an isolated crack,  $c(F)$  varies as  $F^n$ , and such cracks are present at density  $p$  (number per area), the DEF is given by

$$DEF = p \int \left( \frac{F(x,y)}{F_0} \right)^n \, dxdy - 1 \quad (5)$$

The detailed calculation of the DEF is presented for the penny crack + surface reflection in Appendix 3. For  $n=5$ , the DEF is found to be  $8953 \, p a^2 - 1$ , where  $p$  is the surface density of cracks of correct orientation, opening, etc. to produce the field intensification given in Eq. (2). The DEF can be interpreted as  $\Delta N/N$  due to cracks. For  $a \sim 10$  microns and  $m=5$  one needs about 200 appropriate cracks per  $cm^2$  to double the number of damage sites. This density is probably too large to be realistic for high quality optics. Recall it includes only those cracks with the “right” angular orientation to the surface and assumes total internal reflection from the crack wall. Subsurface damage cracks tend to be perpendicular to the surface and crack openings are too small to allow total internal reflection. Since damage initiation is strongly dependent on fluence, reduction in reflection will greatly decrease the DEF. The assumption of S polarization is another selection factor that reduces the actual density  $p$  that should be used.

### Energy absorption in cracks

The above implies that field intensification is an unlikely explanation for the correlation between laser damage initiation and the presence of cracks. We think that enhanced absorption due to cracks is a more plausible explanation.

Cracks are produced during the polishing process when abrasion creates sufficient tensile stress to open cracks. When the stresses are relieved, the cracks close, trapping the polishing slurry, which can contain small absorbing nanoparticles. As an example, we note that ceria is a common ingredient of optical polishing compounds. Removing ceria from the polishing slurry was found<sup>6</sup> to remove the “grey haze” type of damage in fused silica.

Another possibility for enhanced absorption occurs during crack formation. Breaking of material creates fresh free surface in a violent way. As a result, oxygen can escape from  $SiO_2$  creating oxygen deficiency centers (ODC). Oxygen deficiency results in strong UV absorption so clusters of ODC's can form a nanoabsorber. Additionally, the dangling bonds on the crack surface can attract absorbing particles from the environment. It is natural, too, to think that, as a result of friction, the new surface might be electrically charged. Such charges can attract absorbing particles from the environment. Note that absorbing particles

will have more free electrons; hence they have higher polarizability and will adhere to cracks more readily than nonabsorbing particles.

Finally, absorbers can be generated during crack formation. Cracks propagated at high velocity don't move in a straight line. The tip of the moving crack wiggles, chipping off small pieces of material. These small particles have a large surface to volume ratio and therefore a high probability for oxygen to escape. The oxygen deficiency makes them probable small absorbers.

In recent experiments<sup>3</sup>, cracks were controllably produced by application of an indenter in a clean environment. It was demonstrated that the transition from plastic deformation to brittle fracture results in a sharp drop of the damage threshold. The morphology of the damage revealed a chain of localized damage spots along the crack, consistent with the trapped nanoabsorber model.

In summary, we expect that the correlation between cracks and damage is related to small nanoabsorbers (derived from the optical material or the environment) trapped in cracks.

### **Cracking and the extent of damage.**

It is likely that small microabsorbers are the initiators for the observed fused silica surface damage. The laser radiation absorbed in an inclusion and surrounding matrix during the laser pulse generates high pressures, which break the material. All damage takes place long after a nanosecond scale laser pulse terminates and can be treated as the result of a micro-explosion with energy E.

It is clear that the presence of cracks makes the material weaker and increases the extent of the damage which we can estimate using dimensional analysis<sup>7</sup>. There are two types of material modification that occur under high pressure. Hoop stresses around the explosion site fracture surrounding material. Brittle fracture is characterized by the fracture toughness K. According to the Griffith theory<sup>8</sup>, fracture occurs when the stress at the crack tip is high enough that the energy expended in forming new surface area is balanced by the energy gained in releasing strain energy. The stress field near the crack tip varies as  $K'/\sqrt{(x-a)}$  where  $2a$  is the crack length along the x axis. The fracture toughness is the critical value of  $K'$  for which the energy balance occurs. It is determined by Young's modulus Y and surface energy  $\gamma$  as  $(Y\gamma)^{1/2}$  times a numerical constant. The value of K for fused silica is  $0.75 \text{ MPa m}^{1/2}$ . The size of the fracture zone  $R_f$  can be a function of energy E, fracture toughness K and density only. The only possible combination is

$$R_f = \left( \frac{E}{K} \right)^{2/5} \quad (6)$$

Plastic deformation is the second type of modification that occurs. Plastic deformation is characterized by the compressive material strength P with a value of 1.1 GPa for fused silica. The extent of the plastic deformation zone  $R_p$  is determined by E and P and the only dimensionally correct combination is

$$R_p = \left( \frac{E}{P} \right)^{1/3} \quad (7)$$

One sees that for high enough deposited energy, the fracture zone, Eq. (6), is always larger than the plastic deformation zone, Eq. (7). This means a large damage spot must be surrounded by a fracture zone. Small sites correspond to smaller amounts of released energy. In this case, it is difficult to open

cracks, and the damage site consists of only plastic deformation. The energy boundary between these two regimes,  $E_c$ , is given by

$$E_c = \frac{K^6}{P^5} \quad (8)$$

with the size of the damage site being of order

$$R_c = \left( \frac{K}{P} \right)^2 \quad (9)$$

For fused silica, using the material values given above, we find  $E_c \sim 0.1$  nJ and  $R_c \sim 0.5$   $\mu\text{m}$ . The dependence of the two radii on released energy is shown in Fig. (2).

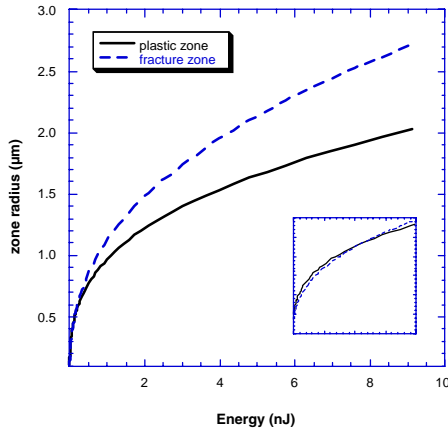


Fig. 2: For released energy of less than 0.1 nJ and size about 1  $\mu\text{m}$ , the plastic zone is larger than the fracture zone (inset). For larger energies, the fracture zone is larger than the plastic deformation zone and cracks extend outward beyond the central plastic zone. The larger the fracture zone, the easier it is to grow the damage with repeated laser exposure. The inset shows crossing of the two sizes at 0.1 nJ.

For fused silica, we expect that small damage spots, of order 1  $\mu\text{m}$ , will not be surrounded by cracks and probably will not grow. Larger craters are surrounded by cracks and much more susceptible to growth. Examples of both kinds of damage spots are shown in Fig. (3).

The above estimates were related to pristine material. In already cracked, i.e. mechanically weakened material the extent of damage will be larger and it will be easier to grow. As a result, inclusions, which are benign in pristine material, can produce observable damage in cracked, weakened material.

## Conclusion

We discussed three ways in which microcracks produced by polishing can affect the damage resistance of optics. Field intensification is possible due to either field discontinuities at crack walls or interference between light reflected from cracks, the output surface and the incoming beam. Quantitative assessment of the amount of intensification implies this is unlikely to be a significant effect for extrinsic damage much less for intrinsic damage. Secondly, cracks can trap absorbing particles resulting either from the polishing slurry or modified silica, e.g. clusters of oxygen deficiency centers. This enhanced absorption

may be responsible for the increased susceptibility to damage. Finally, pre-existing cracks mechanically weaken the material, which leads to more extensive laser damage. Our experience with the improvement of damage resistance with less abrasive polishing (MRF), change in polishing slurry material (ceria) and successful mitigation of damaged sites in fused silica are all consistent with our understanding of the role of cracks.

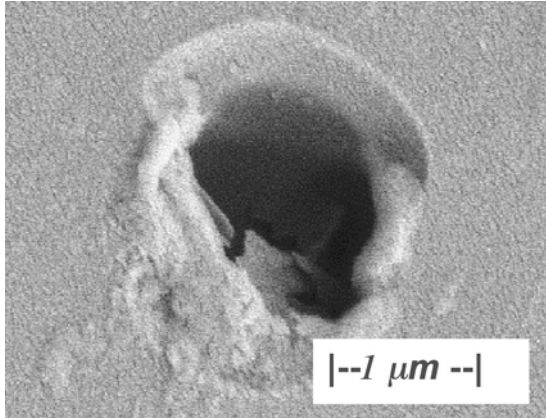


Fig. 3a: Small damage crater in fused silica with smooth walls. Core material has been melted, deformed and partially ejected.

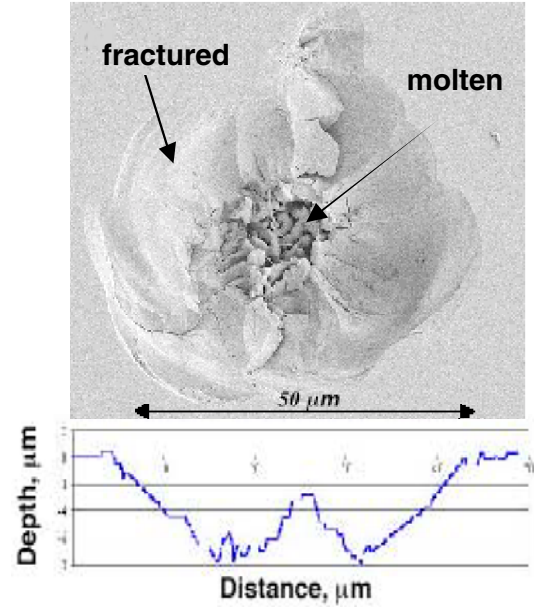


Fig. 3b Large damage crater in fused silica. Core region with remnants of molten and plastically deformed material is surrounded by fractured region.

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## Appendix 1: Intrinsic damage by dielectric breakdown

Free electrons oscillating in the laser field are scattered and gain energy from the electric field at volume rate  $\sigma E^2$ , where  $\sigma$  is the glass conductivity at the laser frequency and  $E$  is the laser electric field. When an electron gains energy larger than the bandgap, 9eV in fused silica, ionization, i.e., generation of new free (conduction band) electrons takes place. For very short and/or very intense pulses, the continuation of this process produces an electron avalanche, and the conduction band electron density grows exponentially in time. The resulting dense electron plasma is highly absorbing and leads to macroscopic damage.

For longer pulses, the Joule heating of electrons is comparable with electron energy losses due to interaction with phonons. If this combined with other losses are greater than the rate at which energy is gained via absorption, the avalanche is quenched. It is difficult to precisely calculate the critical intensity



because the conductivity and, especially, the scattering rate change as a function of electron kinetic energy. Early estimates<sup>9</sup> of the necessary field strength correspond to laser intensity of 60-250 GW/cm<sup>2</sup>. Previously, we used the following arguments to estimate<sup>10</sup> the critical intensity for fused silica. For low intensities, all conduction electrons must have energy near the bottom of the conduction band so we can use the conductivity and scattering rates corresponding to zero energy. The avalanche threshold is given by the condition  $E^2 > \gamma(0)U_{ph}/\sigma(0)$ . For one micron light, this gives an intensity of 80 GW/cm<sup>2</sup>. For  $3\omega$ , using the Drude model for conductivity, we find the threshold intensity to be 2 times higher. Thus, for typical intensities of about 5 GW/cm<sup>2</sup> or less used by NIF, intensification by more than 30 times would be needed to launch an avalanche. There are several caveats to this estimate for ns scale pulses. First other energy loss mechanisms such as thermal conduction and electron diffusion have been neglected. Secondly, the absorption rate drops more than ten times for electrons with energy more than 2eV above the bottom of the conduction band; the conductivity also increases. A definitive estimate would require more careful calculation, but the two above estimates agree in order of magnitude that the intrinsic threshold is much larger than that observed experimentally. These estimates are also consistent with recent measurements<sup>11</sup> on very small defect free volumes, which exhibit high threshold intensities independent of pulselength over a wide range.

## Appendix 2: Damage initiated by small inclusions.

If the intensity of laser light is not sufficient to support an electron avalanche, the damage due to nanosecond pulses usually is initiated by pre-existing defects. The most probable model assumes that the optics subsurface layer contains small absorbing inclusions of different sizes. Laser radiation heats the inclusions to temperatures where the band gap collapses. The radiation is then absorbed in the surrounding material and produces a plasma fireball and subsequent micro-explosion. This damages the material. The detailed picture<sup>12 13</sup> of heating a distribution of small absorbers is consistent with experimental observations, in particular with the observed pulselength dependence of the damage threshold. The model predicts the threshold damage fluence scales as  $\tau^x$ , where  $x \sim 0.3-0.5$  and is mainly due to particles whose size is comparable to the thermal diffusion length  $\sqrt{D\tau}$ . Here D is the substrate thermal diffusivity and  $\tau$  is the laser pulselength.

## Appendix 3: Calculation of DEF

The electric field near the surface for S polarization can be written as follows:

$$E = E_0(1 + 2\cos(k_t x)); \quad F \propto E^2$$

Here  $k_t$  is the component of wavenumber parallel to the rear surface.

The integral in Eq.(5) is given by

$$\int \left( \frac{F}{F_0} \right)^n \frac{dx dy}{S} = \int_0^a (1 + 2\cos k_t x)^{2n} dx / a = \frac{1}{k_t a} \int_0^{k_t a} (1 + 2\cos \xi)^{2n} d\xi$$

We assumed total reflection both from the crack wall and from the surface.

To estimate the number of damage sites due to field intensification assuming that n is an integer, use the binomial expansion of form

$$(1 + 2 \cos[x])^{2n} = \sum_{k=1}^{2n} C_{2n}^k 2^k \cos^k[x]$$

Only the even powers of the cosine survive integration assuming  $k_1 a \gg 1$ . The result is

$$DEF = pa^2 \sum_{k=1}^n C_{2n}^{2k} 2^{2k} \langle \cos^{2k} x \rangle - 1$$

where  $p$  is the crack surface density.

The first few average values of the cosine are presented below:

$$\begin{aligned} \langle \cos^2 x \rangle &= 1/2 \\ \langle \cos^4 x \rangle &= 3/8 \\ \langle \cos^6 x \rangle &= 5/16 \\ \langle \cos^8 x \rangle &= 35/128 \\ \langle \cos^{10} x \rangle &= 63/256 \end{aligned}$$

Finally, we calculate, for  $n=5$ ,  $DEF = 8953pa^2 - 1$ .

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